A promising tool for the investigation of alpha-particle damage in accessory minerals: ⁴He irradiation using a fabricated, Si-based energy filter

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Here we discuss a new idea for how to investigate experimentally the effects of alpha particles (i.e. ⁴He ions with energies in the range 3.9–8.8 MeV) on the structure of accessory minerals. Many accessory species (including zircon, monazite, xenotime, allanite, perovskite, and others) commonly incorporate significant amounts of the radionuclides ²³⁸U, ²³⁵U and ²³²Th. Alpha-decay events in their decay chains cause displacive processes that eventually may create severe structural damage in the host mineral. This is especially the case for recoils of heavy daughter nuclei (0.06–0.16 MeV) upon emission of a high-energy alpha particle: According to cautious estimates, >85 % of the bulk damage is caused by alpha recoils (Nasdala et al. 2001), which create highly localised damage clusters a few ten nanometres in size (Weber et al. 1994).

The effect of the alpha particles themselves, however, is difficult to estimate. Alpha particles transfer most of their energy to the host lattice through so-called ionisation losses. Atomic knock-ons occur predominantly near the end of the He-ion trajectories, where the alpha particles, after being slowed down significantly, may create Frenkel-type defects. The narrow penetration-depth region that suffers significant damage is commonly referred to as Bragg peak; its width lies in the micrometre range. The problem in investigating alpha particle damage by means of MeV He irradiation is that the target material will suffer strong radiation damage only within a correspondingly narrow "stripe" (Fig. 1).



Fig. 1. Cross-polarised light image of a synthetic ZrSiO₄ crystal after irradiation with 1 × 10¹⁶ He ions (8.8 MeV) per cm². The irradiation damage Bragg peak of is seen as narrow "stripe" of lowered birefringence, ca. 32–33 μm below the target surface. For details see Nasdala et al. (2011)

The damage within the Bragg peak ("stripe") is difficult to analyse, for several reasons. Electron-beam techniques with sub-micrometre resolution are critical because the impact of a high-energy electron beam results in damage annealing (Váczi and Nasdala 2017). The application of spectroscopy is problematic also because the damaged layer is shallower than the volume resolution of modern confocal systems (Nasdala et al. 2010, 2011). In addition, the damaged (and hence expanded) "stripe" is sandwiched within less damaged regions; it hence must be affected by strong strain.

A possible way out may be the application of a micromechanically fabricated Si energy filter (Csato et al. 2015). Monte Carlo simulations with SRIM (Ziegler et al. 2010), using the displacement energies of Moreira et al. (2009), visualise the tailoring effect of the energy filter: the sharp Bragg peak is turned into a flat plateau, with a well predictable defect-concentration level in the depth range 23–31 μ m (Fig. 2). We hence propose the conduction of He irradiation of thin zircon foils produced using the focused ion beam (FIB) technique (compare Nasdala et al. 2010), through an Si energy filter and a 26.5±1 μ m zircon window. Concept and first results are presented and discussed.



Fig. 2. a SRIM prediction of the damage distribution in He-irradiated zircon, without and with the use of an energy filter. Note that a 1.5 μ m target (marked red) placed behind a 26.5 μ m zircon window receives well predictable damage. **b** FIB preparation of a 1.5 μ m thick target (SE image)

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References:

- Csato C, Krippendorf F, Akhmadaliev S, von Borany J, Han W, Siefke T, Zowalla A, Rüb M (2015) Energy filter for tailoring depth profiles in semiconductor doping application. Nucl Instrum Meth B 365:182–186
- Moreira PAFP, Devanathan R, Yu J, Weber WJ (2009) Molecular-dynamics simulation of threshold displacement energies in zircon. Nucl Instrum Meth B 267:3431–3436
- Nasdala L, Wenzel M, Vavra G, Irmer G, Wenzel T, Kober B (2001) Metamictisation of natural zircon: accumulation versus thermal annealing of radioactivity-induced damage. Contrib Mineral Petr 141:125– 144
- Nasdala L, Grötzschel R, Probst S, Bleisteiner B (2010) Irradiation damage in monazite (CePO₄): An example to establish the limits of Raman confocality and depth resolution. Can Mineral 48:351–359
- Nasdala L, Grambole D, Götze J, Kempe U, Váczi T (2011) Helium irradiation study on zircon. Contrib Mineral Petr 161:777–789
- Váczi T, Nasdala L (2017) Electron-beam-induced annealing of natural zircon: A Raman spectroscopic study. Phys Chem Miner 44:389–401
- Weber WJ, Ewing RC, Wang L-M (1994) The radiation-induced crystalline-to-amorphous transition in zircon. J Mater Res 9:688–698
- Ziegler JF, Biersack JP, Littmark U (2010) SRIM The stopping and range of ions in matter (2010). Nucl Instrum Meth B 268:1818–1823